

Chemical and Physical Options for Engineering Concrete Performance

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Introduction

Ever since the first concrete bridge, concrete street, and concrete high rise were placed in the U.S. in 1889, 1891, and 1903, respectively, people have attempted to engineer the performance of concrete [1]. As the remainder of this paper will emphasize, desired performance can be engineered through both **chemical** and **physical** means.

In its early days, concrete was a simpler material, usually consisting of only cement, water, sand, and coarse aggregates. Performance-related characteristics (mainly strength) were initially engineered by varying the proportions of these four ingredients, and important parameters such as the water-to-cement ratio (w/c) were soon recognized [2, 3]. Gradually, the important physical characteristics of the three main particulate components of concrete (cementitious materials, sand, and coarse aggregates), such as particle size distribution, relative density, and particle shape were also recognized. While the chemistry of aggregates can be important in specific cases such as alkali reactive aggregates, the chemistry (phase composition) of cements is always influential, particularly in terms of hydration and properties of the hardening concrete.

As with all materials, the performance of concrete is strongly influenced by its physical structure (microstructure), e.g., the arrangement of its component phases within a

three-dimensional space. Parameters such as the volumetric mixture proportions and particle size distributions (PSDs) of the particulate components will strongly influence the initial 3-D structure of the concrete (packing), as well as its properties in the fresh (density, flow properties) and hardened (strength, shrinkage, durability) states. For example, the PSD or fineness of the cementitious materials has been recognized as having a large influence on durability [4], autogenous volume change [5, 6], hydration kinetics [7, 8], setting time and heat release [9], and strength development [10, 11].

From their earliest origins [1], the performance of cement-based materials has been controlled by the judicious addition of supplemental chemicals. For example, the Romans used animal fat, milk, and blood as admixtures to enhance performance of their lime/pozzolana mortars [1]. Over two millennia later, air entraining admixtures were introduced in the 1930s to improve the freeze-thaw resistance of concrete. Currently, concrete designers and technologists have their choice from a nearly endless supply of chemical admixtures including air entrainers, accelerators, retarders, water reducers, superplasticizers, viscosity modifiers, shrinkage-reducing admixtures, corrosion-inhibiting admixtures, admixtures to mitigate alkali-silica reactions, anti-freeze admixtures; the list goes on and on.

It is clear from these examples that from its origins, a dual but parallel track including both physical and chemical approaches to engineering concrete performance has been investigated. In this paper, the engineering of specific properties of concern for the improved performance of modern day concretes will be presented in terms of a comparison of available and potential chemical and physical approaches.

Some Concrete Examples

Rheological Performance

In modern day concretes, the use of water-reducing admixtures and superplasticizers is ubiquitous. These chemicals allow the proportioning of concrete mixtures with w/cm of 0.3 and even lower, leading to the production of higher strength and hopefully higher performance concretes. While there are several classes of chemicals that function admirably in this role, in one way or another, they all lead to a reduction in the interparticle forces between cementitious particles, avoiding the large degree of flocculation that conventionally occurs in a system with only water and cementitious materials. This chemical approach to controlling rheology is quite sensitive to environmental conditions (temperature, etc.), and the retention of adequate rheological properties (yield stress and viscosity) for the several hours that is required to haul, place, and finish a concrete is a challenge. However, the benefits of these materials far outweigh these occasional problems and nearly all concrete mixtures are now proportioned with one or more water-reducing agents/superplasticizers.

The physical PSDs and shapes of both the aggregate and the cementitious material particles also have a strong influence on rheological properties. For example, finer cements are known to have an increased water demand in order to achieve the same flow properties as those of a coarser cement. As another example, fly ash, with its usually spherically-shaped particles, has been noted to improve the rheological properties of pastes, mortars, and concretes in many instances [12]. This may also be due to its effects on interparticle forces and the greater average interparticle spacing of the cement particles in a paste volume “diluted” by fly ash [13]. As pointed out by J. Shilstone Sr. in his comparison of gap-graded and well-graded (Fuller curve) mixtures [14], the grading of aggregates also has a large influence on rheological properties such

as finishability and pumpability. Slump is usually adjusted by changing the dosage of the chemical admixtures. However, in cases where cement-admixture incompatibilities or problems with slump retention are encountered, the volumetric proportions, gradings, and shapes of the particulates comprising the mixture should also be considered and possibly modified to engineer an appropriate and sufficiently robust rheological performance.

Segregation in Self-Consolidating Concrete

As self-consolidating concrete has moved from the laboratory to the field, a problem sometimes encountered in practice is the segregation of the larger aggregates to the bottom of the concrete and the concurrent formation of a paste/mortar rich layer at the top [15]. To avoid this, in addition to increasing the fines content of the concretes, special viscosity modifiers and other chemicals are often added to the mixtures to slow or eliminate the aggregate settlement. Though not as commonly employed, this problem could also be addressed from a purely physical approach by replacing the normal weight coarse aggregates by a pre-wetted lightweight coarse aggregate with a saturated surface dry (SSD) relative density similar to that of the paste component of the concrete. Minimizing the density difference between paste and aggregates minimizes the driving force for the segregation.

Plastic Shrinkage Cracking

From a physical approach, significant reductions in plastic shrinkage cracking have been achieved in recent years with the introduction of plastic (e.g., polypropylene) fibers [16]. For example, at volume fractions as small as 0.2 %, fibers can reduce plastic shrinkage cracking to 10 % of that exhibited by a control system with no fiber addition [16]. Even more recently, a new chemical approach to reducing plastic shrinkage cracking based on the utilization of

shrinkage-reducing admixtures (SRAs) to reduce drying rates and internal stresses has been demonstrated [17, 18].

Autogenous Shrinkage/Early-Age Cracking

Achieving the promise of high performance concrete has been hampered by its increased autogenous shrinkage and accompanying propensity for early-age cracking. Mitigation of autogenous shrinkage can be accomplished by a variety of means, some chemical and some physical in nature [19]. Other than using expansive agents to offset the autogenous shrinkage, the primary chemical means that can be used to reduce autogenous shrinkage is the addition of an SRA to the concrete. Conventionally used to reduce drying shrinkage, the presence of the SRA reduces the surface tension of the pore solution which, in a sealed system, leads to a concurrent reduction in the capillary stresses and strains exerted on the hydrating microstructure [18, 20, 21]. Figure 1 provides an example of the autogenous shrinkage reduction obtained by the addition of 2 % SRA (by mass of cement) to a $w/cm = 0.35$ cement mortar [20]. In this case, the long term autogenous shrinkage is reduced by more than a factor of two.

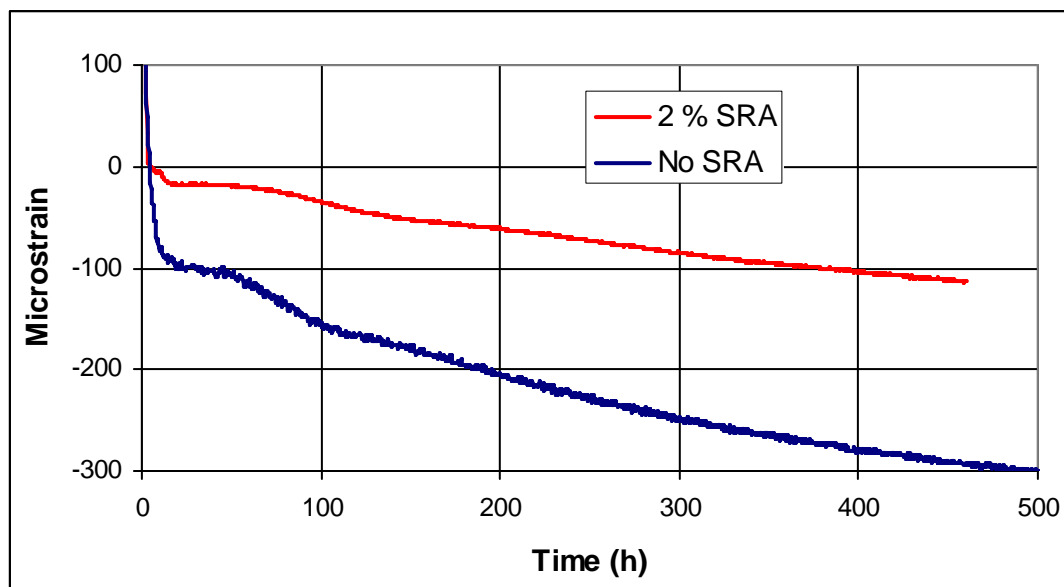


Figure 1. Reduction in autogenous shrinkage (microstrain) obtained by the addition of 2 % SRA to a $w/cm = 0.35$ high performance mortar [20].

There are also several available physical approaches to reducing autogenous shrinkage. For example, as mentioned in the Introduction, autogenous shrinkage is dependent on the fineness of a cement and for a coarse enough cement, an actual autogenous expansion, as opposed to a shrinkage, may be observed at early ages [6]. As an example [6], for a $w/c=0.35$ cement paste, a substantial autogenous shrinkage was observed for the same cement when ground to a Blaine fineness of either $387 \text{ m}^2/\text{kg}$ or $643 \text{ m}^2/\text{kg}$. Conversely, an early age expansion was observed for the two cements produced with Blaine finenesses of $254 \text{ m}^2/\text{kg}$ and $212 \text{ m}^2/\text{kg}$. For these latter two pastes, even after 28 d of sealed curing, the net deformation values were nearly zero, while the finer cements exhibited shrinkages of more than 500 microstrain and evidence of cracking [6]. Of course, a cement with a Blaine fineness of $250 \text{ m}^2/\text{kg}$ is nowhere to be found in the U.S. in the twenty-first century, as most cements are approaching or have exceeded $400 \text{ m}^2/\text{kg}$ [22] (see the high early strength discussion to follow). To the author's knowledge, the coarsest Type I/II cement currently available in the U.S. has a Blaine of about $310 \text{ m}^2/\text{kg}$.

Another physical approach to reducing autogenous shrinkage is the incorporation of internal curing (IC) via the addition of physical internal water reservoirs to the concrete mixture [23, 24]. These reservoirs may consist of pre-wetted lightweight fine aggregates, superabsorbent polymers, or pre-wetted wood fibers. Because they contain "pores" that are larger than those in the hydrating cement paste, water is preferentially drawn from the reservoirs to the paste during hydration. Thus, the small pores in the paste are maintained in saturated conditions and its self-desiccation and the generation of large internal stresses and strains are avoided. The emptying of the larger pores in the IC agent during internal curing does not cause significant shrinkage of the concrete, as the shrinkage stresses are inversely proportional to pore

radius. Figure 2 provides an example of the reduction in autogenous shrinkage that is achievable by the addition of IC agent (with an extra 8 % IC water by mass of cement) to a blended cement mortar. IC is especially important in blended cement systems due to: 1) their generally increased chemical shrinkage relative to conventional cement hydration, which results in an increased demand for additional curing water and 2) their propensity to form a denser, less permeable pore system, such that the efficiency of external water curing can be drastically reduced. For some IC materials, the reservoirs can be employed to distribute chemical admixtures, in addition to water, during the cement hydration process [25].

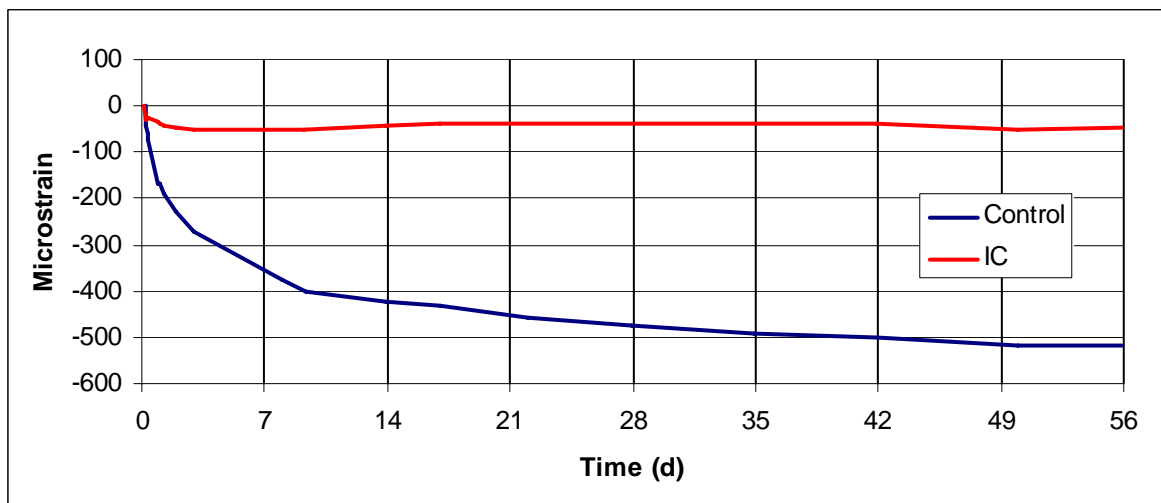


Figure 2. Reduction in autogenous shrinkage (microstrain) obtained by the addition of IC (via pre-wetted lightweight aggregates) to a $w/cm = 0.3$ blended cement mortar containing 20 % slag by mass of cement [24].

High Early Strength

High early strength has been an ever increasing concern of the construction industry in recent years, due mainly to the emphasis on fast-track construction and faster cycle times in the precast industry. In addition to conventional concretes, high early strength formulations have recently been engineered for self-consolidating concretes, precast and repair products, and engineered cementitious composites [26-28]. It has long been recognized that the

physical [10, 11] and chemical properties of the cementitious components of the concrete are critical to strength development, as exemplified by the development of an ASTM standard classification, Type III, for high early strength cement. Since the 1950s, both the fineness and the chemical composition (tricalcium silicate content, alkalis, etc.) of cements have changed substantially, resulting in about a factor of three increase in the 1 day compressive strength of ASTM C109 mortar cubes, for instance [22]. Most cement manufacturers have moved from controlling fineness (Blaine) to controlling the complete PSD of their products. With the newer classifiers, it is now possible to produce a wider range of cement PSDs, such as a cement with basically no particles larger than 30 μm in diameter [29]. With present day technology, cement manufacturers can now (chemically and physically) engineer cements for specific markets or even for specific seasons of the year. Pozzolans such as silica fume are another example of a combined chemical/physical approach to increasing early age hydration and strength; the chemical effects are due to the pozzolanic reactions between the silica fume and the calcium hydroxide produced during cement hydration, while the physical effects are due to the extremely small particle size of a well-dispersed silica fume.

Chemical admixtures have long provided an alternate route to high early strengths. While the original calcium chloride-based admixtures have lost favor due to their negative impacts on corrosion resistance and durability, a variety of non-chloride-based accelerators (calcium nitrates, etc.) have been advanced to quickly fill this role [26-28]. The chemical admixture approach offers the advantage that the dosage (and performance) can be tailored to local environmental conditions that may be changing on a daily basis. If the cementitious components of the concrete are found to be too reactive at early ages, a retarding chemical admixture can also be utilized.

Freeze-Thaw Resistance

As mentioned in the Introduction, air entraining agents were introduced in the U.S. in the 1930s as a chemical means to increase concrete's freeze-thaw resistance. They are employed to stabilize a microscopic system of entrained (by mixing) spherical air bubbles in a fresh concrete. Both the overall air content and the spacing factor that describes how the air bubbles are distributed within the three-dimensional concrete microstructure are critical to assuring adequate freeze-thaw performance [30, 31]. The stability of this chemically-induced air void system during hauling, placement, and pumping has long been an issue, along with the detrimental influences of other components of the concrete (such as fly ashes with a high carbon content) on this stability. A second possibility for obtaining freeze-thaw resistance would be to distribute a "physical" set of air voids within the concrete. For example, pre-wetted lightweight aggregates that are used to provide internal curing may become empty during early-age hydration, and then function successfully as part of an air void system in the hardened concrete [32]. While not conclusively proven by a detailed study, the empirical evidence provided by the excellent durability of lightweight concrete lends credence to this possibility [33]. Superabsorbent polymers used to provide internal curing may also provide a very controlled system of nearly monosize spherical air voids once their work as internal curing agents is completed [34]. Unpublished research has also indicated that other porous materials such as spent fuel cracking catalyst may provide an adequate air void system in concrete. While this "physical" air should be immune to the detrimental influences of other admixtures, fly ashes, etc., it will likely be saturated either prior to its introduction or during its mixing into the fresh concrete, and will thus only function as air voids if this internal water is removed by either self-desiccation or drying

during the hardening process. The spatial and size distributions of these air reservoirs will be of equal importance to their overall volume fraction.

Transport/Reaction-based Degradation Processes

Many degradation processes in concrete require or are significantly enhanced by the ingress of a deleterious species, such as chloride, sulfate, or alkali ions. There are two viable options for mitigating such degradation: 1) providing a physical barrier to the ingress, or 2) using a chemical admixture to avoid, reduce, or alter the degradation (reaction) processes. In the former case, the barrier may be provided by products applied at the concrete surface (sealers, coatings, and crystalline waterproofing products [35]), by products mixed directly into the concrete (hydrophobic admixtures [36]), by the concrete itself (through densification and transport reduction via the use of a low w/cm , a finer cement, or appropriate pozzolans), or at the location of the degradation (epoxy-coated rebars). Examples of the latter approach would include the use of corrosion inhibitors [37, 38] or ASR mitigation admixtures such as lithium-based compounds [39-41].

Computational Tools

In the past ten years, numerous computational tools have been developed and refined for predicting concrete performance as a function of materials and environmental conditions. These tools allow their users to conveniently investigate the influence of many of the chemical and physical approaches described in this article on performance. Table 1 provides the names, web sites, and descriptions of the functionality of a number of **freely** available tools (it is recognized that numerous other tools are available for a fee). Many of these tools have been developed in the Inorganic Materials Group at the National Institute of Standards and Technology; a convenient portal site for all of them can be found at <http://ciks.cbt.nist.gov/cmml.html>.

Tool Name	Web Site	Functionality
HIPERPAV	www.hiperpav.com	HIPERPAV is a user-friendly, Windows based software designed to assess the influence of pavement design, concrete mix design, construction methods and environmental conditions on the early-age behavior of portland cement concrete pavements (PCCP).
Life-365	http://www.corrosioninhibitors.org/life365.htm	Life-365 is a computer program for predicting the service life and life-cycle costs of reinforced concrete exposed to chlorides.
Curing	http://construction.asu.edu/cim/curing/curingfirstpage.htm	This program computes the rate of evaporation for concrete according to nomographs in the American Concrete Institute Standard Practice for Curing Concrete (ACI 308-92). The air temperature, humidity, concrete temperature, and wind velocity are variables which may be known by the user to determine if adequate curing is being achieved.
Internal Curing	http://ciks.cbt.nist.gov/lwagg.html	Resources for internal curing, including mixture proportioning, three-dimensional simulation of water reservoir distribution, and an extensive bibliography.
CIKS	http://ciks.cbt.nist.gov/ciks.html	Computer Integrated Knowledge System for high performance concrete includes mixture proportioning (ACI 211), chloride diffusion, service life prediction, and a virtual rapid chloride permeability test.
COST	http://ciks.cbt.nist.gov/cost	COST (Concrete Optimization Software Tool) is an online design/analysis system to assist concrete producers, engineers and researchers in determining optimal mixture proportions for concrete.
VCCTL	http://vcctl.cbt.nist.gov	Virtual cement and concrete testing laboratory for simulation of hydration and microstructure development, including prediction of numerous physical properties including heat evolution, setting, diffusivities, and strengths.

Summary

“Better concrete through chemistry” is not that far from the actual recent headline and content of the article entitled “**SYNTHETIC CHEMISTRY MOVES INTO CONCRETE**” (Chemical and Engineering News cover story for the Oct. 11, 2004 issue). Chemistry has made and continues to make valuable contributions to endowing concrete with new and desirable properties, increasing durability, and expanding the range of construction applications. Robustness, environmental sensitivity, and detrimental interactions are all issues that will continue to arise as these chemical technologies are applied to variable materials in an even more variable field environment. When these issues do arise, it may be beneficial to consider the possibilities offered by the physical paradigm to engineering concrete performance in place of or in addition to the more commonly employed chemical means.

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